

# Enhancement of Flux Pinning in Neutron Irradiated $\text{MgB}_2$ Superconductor

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$m - H$  loops for virgin and neutron irradiated bulk and powder samples of  $\text{MgB}_2$  were measured in the temperature range  $5 - 30$  K in magnetic field  $B \leq 1$  T. The irradiation at thermal neutron fluences  $9 \cdot 10^{13}$  and  $4.5 \cdot 10^{14} \text{ cm}^{-2}$  caused very small enhancement of  $m - H$  loops at lower temperatures ( $T < 20$  K), whereas the effect at high temperatures was unclear due to difficulty in achieving exactly the same measurement temperature prior and after irradiation. However, the irradiation at  $4.5 \cdot 10^{15} \text{ cm}^{-2}$  produced clear enhancement of  $m - H$  loops (hence  $J_c$ ) at all investigated temperatures, which provides the evidence for the enhancement of flux pinning in  $\text{MgB}_2$  due to ion tracks resulting from  $n + {}^{10}\text{B}$  reaction. The potential of this technique for the enhancement of flux pinning in high temperature superconductors is briefly discussed.

The discovery of new superconductor [1]  $\text{MgB}_2$  aroused large interest in the scientific community [2]. Compared to high temperature superconductors (HTS),  $\text{MgB}_2$  has lower transition temperature,  $T_{co} \cong 39$  K, but its simple composition, abundance of constituents and probable absence of the problems associated with weak intergranular couplings [3,4] (inherent to HTS) make  $\text{MgB}_2$  promising material for the applications in the temperature range  $20 - 30$  K which is still well above  $T_c$ s for low temperature superconductors (LTS). However, compared to LTS employed in the applications of superconductivity,  $\text{MgB}_2$  shows weaker flux pinning which manifests itself in sizably lower critical current density [5]  $J_c$  at 4.2 K. Moreover, flux pinning in  $\text{MgB}_2$  decreases rapidly at elevated temperatures, rendering quite low [6]  $J_c$  for  $T \geq 20$  K. Since as of yet very little effort has been put into optimizing  $J_c$  of  $\text{MgB}_2$ , there is ample space for the improvement of flux pinning in  $\text{MgB}_2$ .

In this respect,  $\text{MgB}_2$  presents almost ideal testground for testing our earlier proposal [7] that flux pinning in boron doped HTS and borocarbides [8] can be enhanced via ion tracks associated with the  $n + {}^{10}\text{B}$  reaction. For these purposes in the case of  $\text{MgB}_2$  one can use  ${}^4\text{He}$  and  ${}^7\text{Li}$  ions from  ${}^{10}\text{B}(n, \alpha){}^7\text{Li}$  reaction. Due to its large cross section at thermal energies ( $\sigma_0 = 3837$  barn) and high abundance of  ${}^{10}\text{B}$  in natural boron (19.78%), one can reach high density of ion tracks in  $\text{MgB}_2$  sample exposed to modest thermal neutron fluences. (This contrasts sharply with U/n and Bi/p treatments employed for the increase of flux pinning in HTS which require high fluences of thermal neutrons and high energy protons respectively [9].) At thermal energies the reaction proceeds only via  ${}^7\text{Li}$  ground and first excited states with the  $\alpha_0/\alpha_1$  branching of 6.7%, i.e. the main contribution comes from the reaction leading to the excited state. In this case the reaction products  ${}^4\text{He}$  and  ${}^7\text{Li}$  nuclei have energies of 1.47 and 0.84 MeV respectively. The sum of their ranges in  $\text{MgB}_2$  with  $\rho = 2.6 \text{ gcm}^{-3}$  is about  $6 \mu\text{m}$ .

Large cross section  $\sigma_0$  for  ${}^{10}\text{B}(n, \alpha){}^7\text{Li}$  reaction and high boron content in  $\text{MgB}_2$  poses problems in irradiation of bulk samples. The mean free path of thermal neutrons in  $\text{MgB}_2$  is about 0.2 mm. Obviously, in samples with thickness of  $\sim 1$  mm the ion tracks will be very unevenly distributed. In an isotropic neutron field more than half of the ion tracks would be in the first 100  $\mu\text{m}$  from the surface. The problem is somewhat facilitated in thinner samples, but the homogenous distribution of ion tracks in the samples is still unlikely.

The simplest way to monitor the enhancement of flux pinning (an increase in  $J_c$ ) in type II superconductors is to measure the magnetization hysteresis curves ( $m - H$  loops). Since  $J_c$  is proportional to irreversible magnetization, any increase in the breadth of  $m - H$  loop shows the enhancement of flux pinning. This method can also be applied to powder samples where more direct transport measurements of  $J_c$  are not possible. In what follows we present the preliminary results of magnetization study performed on virgin and neutron irradiated bulk and powder samples of  $\text{MgB}_2$ . Although these results were obtained for rather thick samples ( $d \geq 1$  mm) and quite low neutron fluences, they indicate an enhancement of flux pinning following neutron irradiation both in bulk and powder samples. Bulk  $\text{MgB}_2$  samples were cut from pellet prepared by conventional solid state reaction [10]. SEM studies revealed coarse grained structure with grain size of order [10] of 200  $\mu\text{m}$ . The measured density was  $\sim 1 \text{ gcm}^{-3}$ , i.e. about 0.4 of the ideal density of  $\text{MgB}_2$ . The sample for magnetization study had dimensions  $3.8 \times 1.5 \times 1 \text{ mm}^3$  and mass of 5.93

mg. Similar sample (cut from adjoining part of the pellet) was used for ac susceptibility study of superconducting transition temperatures prior and after neutron irradiation. The powder sample was prepared by mixing small amount of commercial (Alfa Aesar)  $\text{MgB}_2$  powder (325 MESH size) with about three times larger volume of epoxy in a plastic pill. After setting, plastic container was removed and cylindrical sample with diameter 5.2 mm and length about 5 mm was split into two approximately semicylindrical samples used for magnetization and ac susceptibility measurements respectively. The  $m - H$  loops were measured with commercial vibrating sample magnetometer (VSM) in magnetic field up to 1 T at field sweep rate 15 mT/s. Ac susceptibility was measured with highly sensitive ac susceptometer [11]. The employed temperature range was 5 – 40 K. The samples were irradiated at the roundabout of the TRIGA Mark II reactor of the Jožef Štefan Institute in Ljubljana. At reactor power of 25 kW the rotational irradiation facility has a flux of  $1.45 \cdot 10^{11} \text{ cm}^{-2}\text{s}^{-1}$  thermal ( $E < 0.5 \text{ eV}$ ) and  $0.22 \cdot 10^{11} \text{ cm}^{-2}\text{s}^{-1}$  fast ( $E > 0.1 \text{ MeV}$ ) neutrons. Initial thermal neutron fluence for all samples was  $9 \cdot 10^{13} \text{ cm}^{-2}$ . In later experiments, we employed fivefold and fiftyfold larger thermal neutron fluences.

The ac susceptibility of bulk virgin sample (Fig. 1) showed superconducting transition with diamagnetic onset at  $T_{co} = 38.2 \text{ K}$  and transition width  $\Delta T_c(0.1 - 0.9) = 0.46 \text{ K}$  in ac field amplitude of 1.5 A/m. In spite of its porosity, sample showed excellent grain connectivity as manifested by smooth, single step transitions at elevated ac fields ( $\sim 10^4 \text{ A/m}$ ). The comparison of low field–low temperature diamagnetism in our sample with that in niobium sample of approximately the same shape indicated Meissner fraction  $\geq 80\%$ . The powder sample (Fig. 1) showed similar  $T_{co} = 38.2 \text{ K}$  (inset to Fig. 1), but most of transition was shifted to lower temperature compared to that for bulk sample. At low field (1.5 A/m) this shift was 0.9 K at half transition, and the transition showed shallow tail below 35 K which persisted down to 4.2 K (the transition in bulk sample was completed at 35 K). These phenomena are consequence of a broad grain size distribution (grain sizes  $\leq 43 \mu\text{m}$ ) and impurity content in commercial  $\text{MgB}_2$  powder. Both for powder and bulk samples, the superconducting transitions after irradiations to  $9 \cdot 10^{13}$  and  $4.5 \cdot 10^{14} \text{ cm}^{-2}$  remained practically the same as those for virgin samples. In particular,  $T_{co}$  remained within 0.1 K of the initial value and the breadths and magnitudes of the diamagnetic transitions practically did not change. However, neutron fluence of  $4.5 \cdot 10^{15} \text{ cm}^{-2}$  caused small shift of diamagnetic transition ( $\approx 0.3 \text{ K}$  at half transition) towards lower temperature for bulk sample (Fig. 1).

Fig. 2 shows magnetic moment vs. applied field loops for bulk sample at 5 and 10 K. The results for both virgin and irradiated (dashed line) sample are shown. The field sweep rate was 15 mT/s. At elevated fields the irradiation seems to produce marginal increase in the irreversible magnetic moment and this increase is somewhat more pronounced at 10 K than at 5 K. This result is consistent with rather low employed neutron fluence, uneven distribution of ion tracks in our thick sample (most tracks concentrated within thin surface layers) and much stronger (dominant) pinning effect of intrinsic pinning centres which manifests itself in high critical current densities of bulk  $\text{MgB}_2$  samples at low temperatures [5].

However, the reappearance of the partial flux jumps (manifested as sharp roughly zig-zag variation of magnetic moment at lower field in Fig. 2) in irradiated sample at 10 K (note that virgin sample did not show flux jumps at 10 K) seems to support the enhancement of flux pinning at 10 K after neutron irradiation. These partial (pseudo) flux jumps have already been reported for bulk  $\text{MgB}_2$  samples similar to our one [10] at low temperatures ( $T < 10 \text{ K}$ ). In contrast to normal flux jumps in LTS [12], the above phenomenon is associated with violent flux entry in the surface layers of porous sample. The phenomenon occurs only at low temperatures where intragranular flux pinning greatly exceeds that in intergranular links. Therefore, the reappearance of this phenomenon upon irradiation seems to indicate an increase in the intragranular pinning associated with ion tracks.

The  $m - H$  loops for both virgin and neutron irradiated ( $9 \cdot 10^{13} \text{ cm}^{-2}$ ) powder sample were measured at temperatures 5, 10, 20 and 30 K, respectively. For  $T = 5$  and 10 K the  $m - H$  loops of irradiated powder were identical to those of virgin one, whereas at 20 and 30 K  $m - H$  loops of irradiated sample showed small and sizable enhancement, respectively. No effect of light irradiation on  $m - H$  loops of  $\text{MgB}_2$  powder for  $T \leq 10 \text{ K}$  was plausible. It probably arises from combination of strong (dominant) intrinsic flux pinning in  $\text{MgB}_2$  powders (manifested by an order of magnitude larger  $J_c$ s of  $\text{MgB}_2$  powders compared to those of corresponding bulk samples [3,13]) and employed low neutron fluence (hence low ion track density). However, large increase in the breadth of  $m - H$  loops of irradiated powder at 30 K was unexpected. Although the intrinsic flux pinning in  $\text{MgB}_2$  grains decreases rapidly at elevated temperatures [6], the pinning by ion tracks also becomes less efficient due to corresponding increase of the coherence length of  $\text{MgB}_2$  [5] with temperature. Therefore, large effect of low neutron fluence on  $m - H$  loops at 30 K seems unlikely.

Furthermore, rapid variations of  $J_c$  with temperature for  $\text{MgB}_2$  samples at elevated temperatures [3,5,6,10,13] makes the comparison of high temperature  $m - H$  loops obtained in two experiments reliable only if the sample temperatures were practically identical in two experiments. This condition is difficult to fulfill with the temperature control technique commonly used for VSMs. In VSM preheated helium gas flows around sample and there is no thermometer at the sample holder. Therefore, the actual sample temperature depends somewhat on the combination of the helium flow rate, duration of the temperature stabilization and the size and properties (such as shape, thermal

capacity and conductivity, etc.) of the sample. Because of this, we performed additional experiments in order to ascertain the role of ion tracks in flux pinning in  $\text{MgB}_2$ . In the second experiment we irradiated two powder and one bulk  $\text{MgB}_2$  sample to five times larger neutron fluence ( $4.5 \cdot 10^{14} \text{ cm}^{-2}$ ) than that used in first experiment and measured their  $m - H$  loops prior and after irradiation at temperatures 10, 20 and 30 K. In order to minimize the eventual difference in sample temperature in subsequent measurements we prepared plate-like powder samples with thickness  $d \approx 1 \text{ mm}$  and used longer time intervals for the temperature stabilization ( $t_s \geq 15 \text{ min}$ ).

Fig. 3 shows  $m - H$  loops both for virgin and irradiated ( $4.5 \cdot 10^{14} \text{ cm}^{-2}$ )  $\text{MgB}_2$  powder at temperatures 10, 20 and 30 K, respectively. The breadth of  $m - H$  loop after irradiation is slightly larger at 10 K, shows almost no change at 20 K and appears somewhat smaller at 30 K. The results for other powder sample were practically the same. Whereas the results for 10 K and 20 K can be interpreted in terms of decreasing efficiency of ion track pinning on increasing temperature, the decrease of  $m$  (hence  $J_c$ ) of irradiated sample at 30 K is at variance with the fact that irradiation produced no measurable change in either transition temperature or the shape of transition for  $\text{MgB}_2$  powders. No change in superconducting parameters of the samples is difficult to reconcile with the suppression of their  $J_{cs}$  at 30 K. The most probable explanation of this observation (Fig. 3) is that our control of the sample temperature, although improved, is still insufficient for the reliable measurements of small changes in  $J_c$  at 30 K. The  $m - H$  loops of bulk  $\text{MgB}_2$  sample irradiated to the same neutron fluence was at 20 K almost unchanged compared to that for virgin one, whereas at 30 K it showed small enhancement. No enhancement of  $m - H$  loop at 20 K probably indicates that the enhancement at 30 K was at least partially due to a slightly lower measurement temperature after irradiation. However, the  $m - H$  loops of irradiated sample showed strong flux jumps at 10 K (which were not observed in virgin sample) thus clearly indicating an enhancement of flux pinning (larger  $J_c$  [14]) after irradiation.

Taken together, these experiments show small enhancement of flux pinning at lower temperatures ( $T < 20 \text{ K}$ ) in irradiated  $\text{MgB}_2$  samples (both bulk and powder ones). However, for the employed (low) neutron fluences the effects are too small to allow more quantitative assessment. Therefore, we performed third experiment in which two powder and one bulk  $\text{MgB}_2$  sample were irradiated to neutron fluence  $4.5 \cdot 10^{15} \text{ cm}^{-2}$ . After irradiation the superconducting transition of bulk sample was shifted a little towards lower temperature, the shift at half transition being about 0.3 K (Fig. 1). Since the powder samples showed an induced radioactivity, probably caused by some impurity present in commercial  $\text{MgB}_2$  powder (95.5% purity), the measurements on these samples were postponed.

Fig. 4 shows  $m - H$  loops both for irradiated and virgin bulk  $\text{MgB}_2$  sample at temperatures 10, 20, 25 and 30 K, respectively. The appearance of flux jumps at 10 K and the increase in the breadths of  $m - H$  loops for irradiated sample at all other temperatures prove the enhancement of flux pinning after irradiation. We also observe that the enhancement of  $J_c$  for  $T \geq 25 \text{ K}$  decreases with increasing temperature, which is partially due to the decrease of  $T_c$  upon irradiation (Fig. 1). The enhancement of  $J_c$  due to ion track flux pinning at 20 K is about 2% and 5% at  $\mu_0 H = 0$  and 0.6 T, respectively. Clearly, the enhancement of  $J_c$  will become larger at higher applied fields. The observed rather modest enhancement of  $J_c$  in irradiated bulk  $\text{MgB}_2$  sample is probably a consequence of quite large coherence length in  $\text{MgB}_2$ , small masses and energies of ions and the use of still rather modest neutron fluence. Since in spite of porosity of our sample the decrease of  $T_c$  is quite small, the use of sizably larger neutron fluences seems feasible, which would result in correspondingly larger enhancement of flux pinning. The other advantage of flux pinning by ion tracks resulting from  $n + {}^{10}\text{B}$  reaction is that  $J_c$  is enhanced at all fields (including  $H = 0$ , Figs. 3 and 4), whereas the other ion tracks invariably suppress  $J_c$  at  $H = 0$  and the enhancement of  $J_c$  occurs only at elevated fields [9]. Furthermore, the enhancement of flux pinning by ion tracks from  $n + {}^{10}\text{B}$  reaction in HTS (such as Bi-Sr-Ca-Cu-O compounds) should be sizably larger due to considerably smaller coherence lengths and weaker intrinsic flux pinning in these materials. Taken together, our results confirm the flux pinning effect of ion tracks in  $\text{MgB}_2$  and moreover enable us to predict that the same technique may become powerful method for the enhancement of flux pinning in HTS [7].

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FIG. 1. Variations of real ( $\chi'$ ) and imaginary ( $\chi''$ ) part of ac susceptibility for commercial  $\text{MgB}_2$  powder embedded in epoxy (dashed) and bulk  $\text{MgB}_2$  sample prior (full) and after irradiation at thermal neutron fluence  $4.5 \cdot 10^{15} \text{ cm}^{-2}$  (dotted) with temperature for alternating field amplitude  $H_m = 15.8 \text{ A/m}$ . The inset: enlarged view of the onset of superconductivity for the same samples.

FIG. 2. Magnetization loops for bulk  $\text{MgB}_2$  sample at 5 and 10 K. Dashed line denotes results obtained after irradiation of sample to thermal neutron fluence of  $9 \cdot 10^{13} \text{ cm}^{-2}$ .

FIG. 3. Magnetization loops for  $\text{MgB}_2$  powder embedded in epoxy at 10, 20 and 30 K, respectively. Dashed line is for the same sample exposed to neutron fluence of  $4.5 \cdot 10^{14} \text{ cm}^{-2}$ .

FIG. 4. Magnetization loops for bulk  $\text{MgB}_2$  sample at 10, 20, 25 and 30 K, respectively. Dashed line is for the same sample exposed to the neutron fluence  $4.5 \cdot 10^{15} \text{ cm}^{-2}$ .









